# SYNTHESIS OF CYCLODEXTRIN (CD) USING IMMOBILIZED ENZYME ON HOLLOW FIBER MEMBRANE

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MASTER OF SCIENCE

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We hereby declare that we have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science.

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I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

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# SYNTHESIS OF CYCLODEXTRIN (CD) USING ENZYME IMMOBILIZED ON HOLLOW FIBER MEMBRANE

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## ABSTRAK

Siklodekstrin (CD) dihasilkan daripada tindak balas enzimatik siklodekstrin glukanotransferase (CGTase) di dalam kehadiran kanji sebagai substrat. CD mempunyai keupayaan untuk bertindak sebagai ejen perangkum dan juga aditif yang digunakan di dalam pelbagai aplikasi. Walau bagaimanapun, penggunaan CGTase selalunya terhad disebabkan ketidakstabilan enzim dan mudah denaturasi semasa penghasilan CD. Justeru, enzim imobilisasi telah dikaji secara meluas bagi mengatasi masalah-masalah ini. Pelbagai kaedah dan penyokong untuk imobilisasi telah dikaji selama bertahun-tahun, tetapi masih kurang kajian secara mendalam tentang kesan reaksi parameter-parameter untuk penghasilan CD menggunakan imobilisasi CGTase. Objektif kajian ini adalah untuk menentukan dan mengoptimumkan kesan reaksi parameter-parameter untuk penghasilan CD menggunakan CGTase yang diimobilisasi pada poliviniliden fluoride membran gentian berongga. Kesan jenis kanji, kepekatan kanji, suhu, pH, masa reaksi dan kadar agitasi pada penghasilan CD telah dikaji dengan menggunakan kaedah satu faktor pada satu masa (OFAT). Hasil kajian menunjukkan bahawa 3% (w/v) kepekatan larutan kanji ubi kayu telah menghasilkan jumlah CD yang paling tinggi (5.22 mg/mL). Penghasilan CD juga merupakan yang paling tinggi pada suhu 40°C (5.21 mg/mL), pH 6 (4.62 mg/mL), 6 jam masa reaksi (4.37 mg/mL) dan 150 rpm agitasi (4.38 mg/mL). Berdasarkan keputusan yang diperoleh, imobilisasi CGTase menghasilkan CD 2 ke 3-kali ganda lebih banyak berbanding enzim bebas. Penyaringan parameter-parameter yang signifikan untuk penghasilan CD telah dijalankan dengan menggunakan reka bentuk faktorial pecahan (FFD). Hasil kajian menunjukkan kepekatan larutan kanji ubi, pH dan suhu adalah parameter yang paling signifikan. Parameter-parameter tersebut kemudiannya dioptimumkan dengan menggunakan reka bentuk komposit pusat (CCD). Dengan menggunakan keadaan yang optimum (3.2% w/v kepekatan larutan kanji ubi kayu, 45.2°C suhu reaksi dan pH 5.6), penghasilan CD adalah 3.2-kali ganda lebih banyak berbanding sebelum proses pengoptimuman. Kestabilan haba dan pH lebih luas telah ditunjukkan oleh imobilisasi CGTase berbanding enzim bebas. Imobilisasi enzim telah berjaya mengekalkan sehingga 38% daripada aktiviti awal dan boleh digunakan semula sebanyak 10 kali untuk penghasilan CD. Oleh itu, imobilisasi CGTase menggunakan membrane gentian berongga membuktikan bahawa ianya sesuai untuk meningkatkan penghasilan CD dengan kestabilan enzim yang tinggi.

## ABSTRACT

Cyclodextrin (CD) is produced by the enzymatic reaction of cyclodextrin glucanotransferase (CGTase) in the present of starch as a substrate. The CD has the ability to act as encapsulating agent and also additives in many applications. However, the used of CGTase is often limited due to the instability of the enzyme and easily denatured during the production of CD. Hence, enzyme immobilization has been widely explored to overcome these problems. Various immobilization methods and supports have been investigated over the years, yet less study was conducted in detail on the effect of reaction parameters for the production of CD by using immobilized CGTase. The objectives of this study were to determine and to optimize the effect of reaction parameters for the production of CD by using the immobilized CGTase on polyvinylidene difluoride (PVDF) hollow fiber membrane. The effect of starch type, starch concentration, temperature, pH, reaction time and agitation rate on the CD production were determined by using one factor at a time (OFAT). The results revealed that 3% (w/v) of soluble potato starch produced the highest amount of CD (5.22 mg/mL). The production of CD was also the highest at temperature of 40°C (5.21 mg/mL), pH 6 (4.62 mg/mL), 6 h of reaction time (4.37 mg/mL) and 150 rpm of agitation rate (4.38 mg/ml). From the result, immobilized CGTase managed to produce 2 to 3-fold higher of CD production than the free CGTase. The screening of the significant reaction parameters for the production of CD were conducted by using fractional factorial design (FFD). The results showed that the concentration of soluble potato starch concentration, pH and temperature were the most significant parameters on the production of CD. The reaction parameters were then optimized by using central composite design (CCD). Under the optimized conditions (3.2% w/v concentration of soluble potato starch, 45.2°C of reaction temperature and pH 5.6), the production of CD was 3.2-fold higher than the amount of CD before the optimization process. The immobilized enzyme was successfully retained up to 38% of the initial activity and can be reused for 10 cycles for the production of CD. Therefore, the immobilization of CGTase on hollow fiber membrane proved to be suitable to enhance the production of CD with high enzyme stability.

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## LIST OF SYMBOLS

| | |
|---|---|
| $\alpha$ | Alpha |
| $\beta$ | Beta |
| $\gamma$ | Gamma |
| $K_m$ | Michaelis-Menten |
| S | Substrate concentration |
| V | Initial production rate |
| $V_{max}$ | Maximum velocity |
| $M_w$ | Molecular weight (g/gmol) |
| $D_f$ | Dilution factor |
| w/v | Weight solute per volume solution |
| °C | Degree Celsius |
| min | Minutes |
| hr | Hour |

## LIST OF ABBREVIATIONS

| | |
|---|---|
| ANOVA | Analysis of variance |
| BBD | Box-behnken design |
| CCD | Central composite design |
| CD | Cyclodextrin |
| CGTase | Cyclodextrin glucanotrasferase |
| FESEM | Field emission electron microscopy |
| FFD | Full factorial design |
| HCl | Hydrochloric acid |
| HPLC | High Performance Liquid Chromatography |
| NaOH | Sodium hydroxide |
| MW | Molecular weight |
| OFAT | One factor at a time |
| PVDF | Polyvinylidene fluoride |
| rpm | Revolution per minute |
| RSM | Response surface methodology |
| sp. | Species |

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